

## **Measurements of the Aerosol Light-Scattering Coefficient at Ambient and 85% Relative Humidity on the ONR Pelican During ACE-2**

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### **LONG-TERM GOALS**

Associated with the renewal of funding in 2002, and the successful attainment of several of the studies early goals, the current goals of the study have been modified and now include the interaction of marine aerosols with marine boundary layer (MBL) clouds. New goals include: 1) assessment of the relative impacts of purely meteorological and aerosol factors on cloud albedo, 2) determination of the impact of very large CCN on cloud albedo, 3) assessment of the impact of cloud processing on aerosol light scattering, and 4) explore, observationally, the impact of organics on CCN activation in MBL clouds. Additionally, the more general goal of assessing the impact of organics on aerosol hygroscopicity has been retained and, indeed, expanded to include an element of aerosol-cloud interaction. On the other hand, we have curtailed our exploration of the hygroscopicity-organic composition relationship using the POEM code since our work suggests that it is premature to attempt this until measurement of the aerosol size distribution in the super-micron size range is better established.

### **OBJECTIVES**

During the current year, we have concentrated primarily on analysis of data gathered during the CARMA-III campaign. We have also begun to collate data from this campaign with previously acquired data from CARMA-II. Our specific objectives for the past year are as follows.

- Analysis of aerosol hygroscopicity over a broad size-range using Aerosol Hydration Spectrometer (AHS) data acquired during CARMA-III.
- Explore the impacts of aerosol chemical properties on CCN concentration and cloud drop number concentration (CDNC) in marine stratocumulus.

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- Explore the impact of aerosols on cloud albedo and compare this with the impact of such purely meteorological parameters as the Sea Surface Temperature (SST) utilizing data from both CARMA-II and CARMA-III.

## **APPROACH**

Obtaining reliable measurements of both super-micron aerosol particles and their hygroscopicity is a major goal of our program. Our primary tool for this goal is the AHS described in last years annual report, coupled with the Descriptive Hygroscopic Growth Factor analysis technique also described in that report. For the association of the measured hygroscopicity as a function of size with chemical composition, we have used several different substrate-based approaches for measuring the aerosol chemistry. Most novel is the use of the Pacific Northwest National Laboratory (PNNL) TRAC sampler. This device is essentially an automated SEM/TEM substrate sampler that can expose substrates as frequently as every 30 s. The substrates are analyzed post-flight by a variety of powerful analytical techniques (cf., Laskin et al, 2003; Hoffman et al, 2004). These techniques can yield size-resolved chemical composition data to collate with the size-resolved hygroscopicity data from the AHS. However, the TRAC sampler had not been deployed in a marine venue prior to the CARMA-III study and some sampling issues arose which sharply limited the amount of useful data that could be collected (discussed in results section).

To address the second goal, a combination of field measurements of below cloud aerosol size distribution and composition was used to initialize a kinematic parcel model with size-resolved chemistry and microphysics. The model predicted the CDNC in the overlying cloud, a prediction then compared to direct measurements of the CDNC in a closure analysis.

The methodology for achieving the third objective involves the use of satellite retrievals of albedo ( by both MODIS, AVHRR and GOES) to compare with in situ measurements of cloud properties and below cloud aerosol and SST gradients. The instruments to measure the in situ parameters are part of the standard instrument package on the CIRPAS Twin Otter and have been previously described.

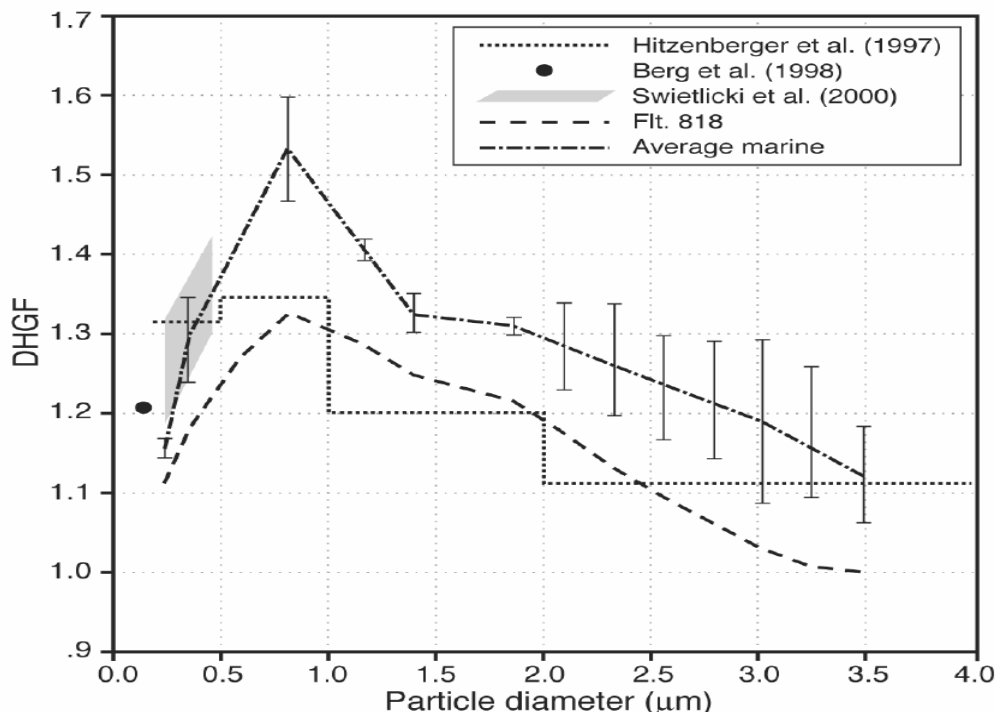
## **WORK COMPLETED**

Data have been obtained from CARMA-III on the size resolved hygroscopicity of marine aerosol and its relationship to composition. An analysis of the relationship between various DHGF spectra and aerosol type has been completed and is in the review process. A second analysis of the impact of aging on DHGF spectra and also an analysis of possible kinetic limitations to hygroscopic growth is well underway. A closure study of the aerosol size and composition relationship to CDNC has been completed and the result published. Finally, sufficient data have been obtained from CARMA-II and CARMA-III to permit an assessment of the relative impact of mesoscale aerosol gradients and various other parameters on cloud albedo variations over the same scale. This analysis has been accepted for publication.

## **RESULTS**

Data addressing our first goal were gathered during CARMA-III. And shown in Figure 1. The average marine spectrum of DHGF's shown in the figure displays a shape characteristic of virtually all of the background marine aerosol samples, though, as suggested by the error bars, there was considerable sample to sample variability, particularly at the larger sizes. The peak in hygroscopicity between about

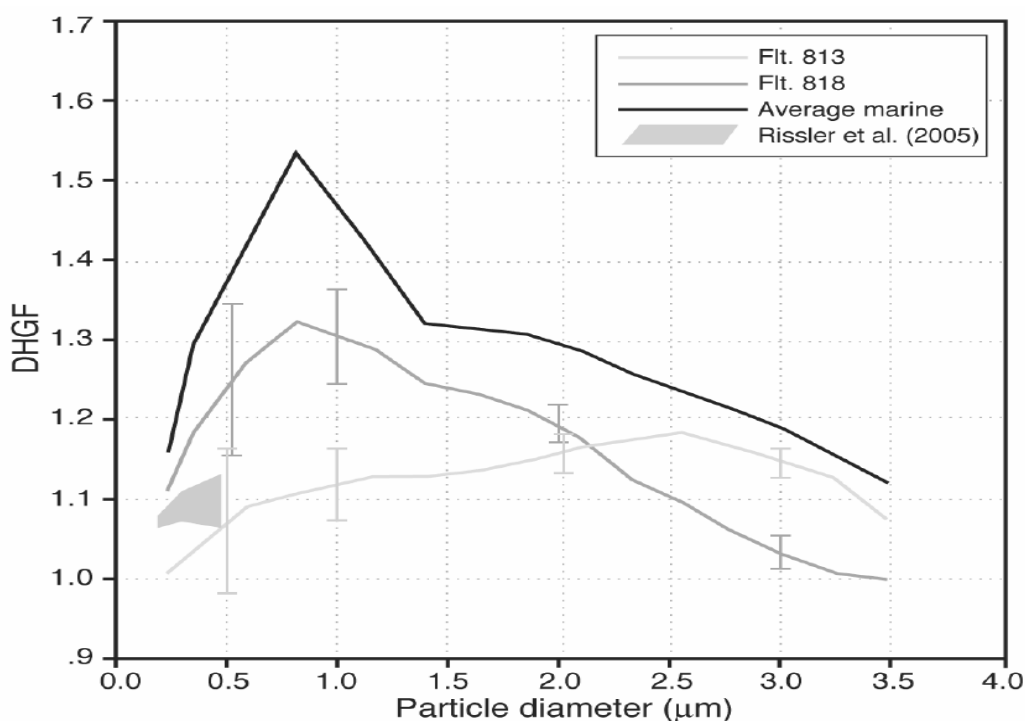
0.5 and 1.5  $\mu\text{m}$  diameter, while consistent with previous studies as indicated in the figure, is an important feature that becomes clear only within the context of the broad range of our measurements.



**Figure 1. Mean marine DHGF spectrum measured during CARMA-III and a polluted aerosol DHGF spectrum measured August 18<sup>th</sup> (flt 818) compared to selected previous measurements. The previous measurements have been adjusted to the AHS measurement RH range using the hygroscopicity determined in these studies.**

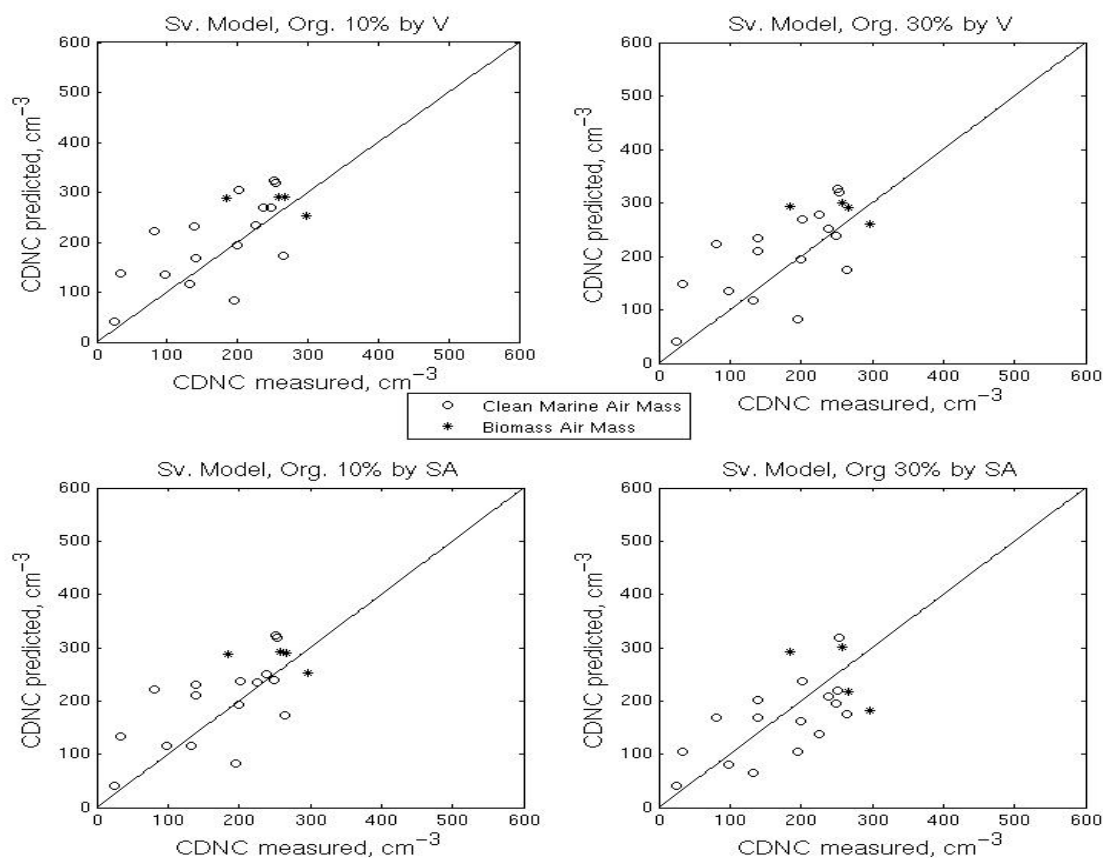
It is noteworthy that the polluted aerosol spectrum has a shape very similar to that of the marine spectra but is systematically lower. It does, however, compare very favorably with the measurements of Hitzenberger et al (1997), taken in Mainz under obviously polluted conditions but utilizing aerosol on filters hydrated in an environmental chamber to determine hygroscopicity. The comparison of the marine and polluted spectra implicitly supports the dependence of hygroscopicity on aerosol composition.

A more extreme example of such a compositional dependence is shown in Figure 2. In this figure the average marine spectrum and the polluted spectrum from Figure 1 are now contrasted with a spectrum from a forest fire plume advected out to sea from a fire in southern Oregon. The HYSPLIT back trajectory for this sample suggests a travel time of  $\sim 24$  hrs. The most striking difference between this fire aerosol sample and the others is the absence of the peak in hygroscopicity around 1  $\mu\text{m}$  diameter. Tests with the enhanced delay time option of the AHS suggest that this is in part ( $\sim 15\%$ ) a kinetic phenomenon, i.e., that the fire aerosol is simply slow to reach thermodynamic equilibrium. However, it is mostly associated with a real difference in water equilibrium for this aerosol. In this regard, it is important to note that the fire aerosol is  $\sim 80\%$  organic, a much higher percentage than any of our other samples. Comparison with data from the recent study by Rissler et al(2005) suggest that this low hygroscopicity is may be a common characteristic of fire aerosols.



**Figure 2. Comparison of the DHGF spectrum from a biomass fire (flt 813) with the polluted spectrum from flt 818 and the average marine spectrum. Error bars are 95% confidence intervals.**

Cloud condensation nuclei closure was examined with the data collected from the CARMA III (Cloud and Aerosol Research in the Marine Atmosphere) campaign conducted off the coast of Monterey, California during August, 2005, to determine the impact of organics on aerosol activation. Cloud base conditions and below cloud aerosol size-number concentrations were measured during the campaign and used to initialize an adiabatic, kinematic cloud parcel model with basic chemistry. The most adiabatic cases were chosen for closure studies, as specified by Yum and Hudson (2002). The model underwent several modifications to emulate the potential impacts of organics on aerosols in the supersaturated environment, and the modeled results were compared to measured cloud droplet number concentrations (CDNC). The organics were modeled as an insoluble core, surfactants, and with explicit solution activity, in concentrations ranging from 10 – 30% of the total aerosol mass. Some results from these closure calculations are shown in Figure 3

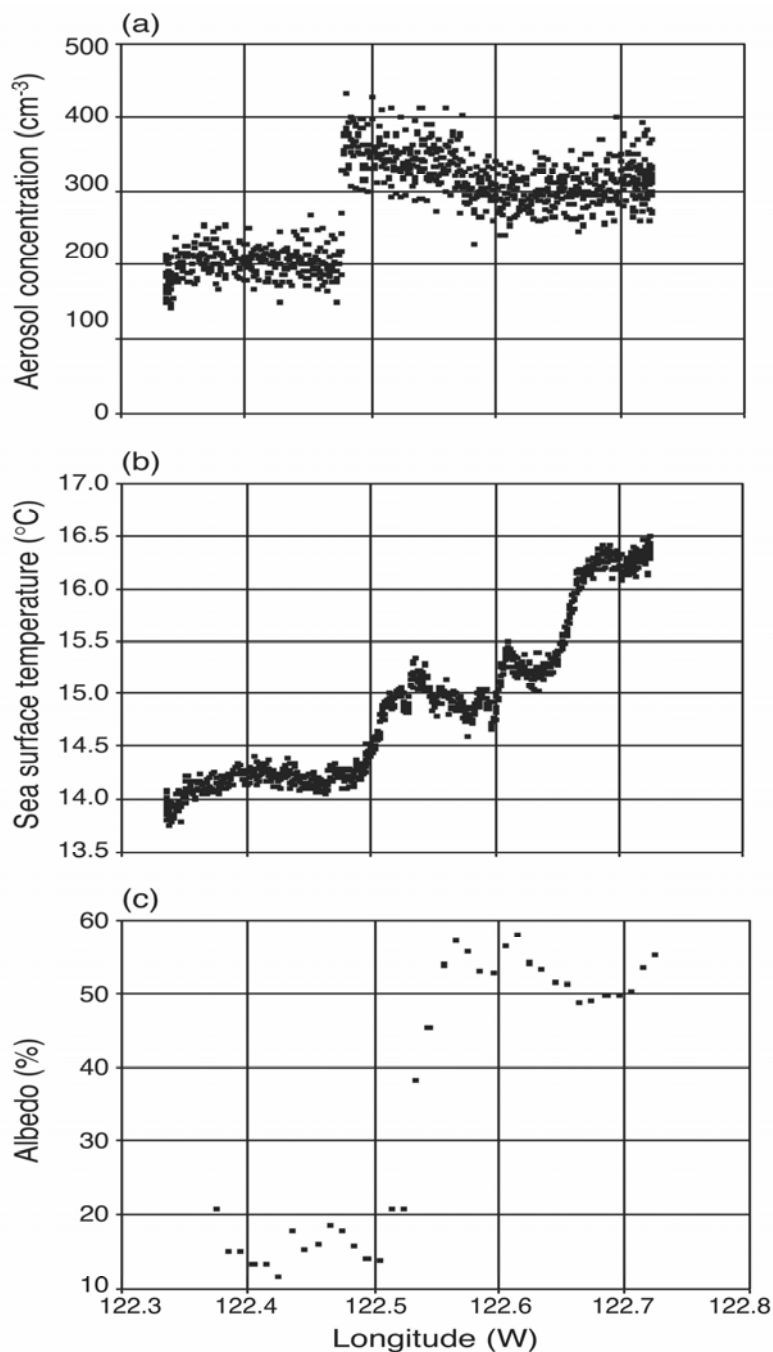


**Figure 3. The agreement between the predicted and measured CDNC using nonideal water activity calculations. The models use the Svenningsson calculations (Sv.) of solution activity with 10-30% organics distributed by volume (V), and surface area (SA). The air masses that contain significant biomass burning aerosol are indicated, and the one-to-one line is also included in the graphs.**

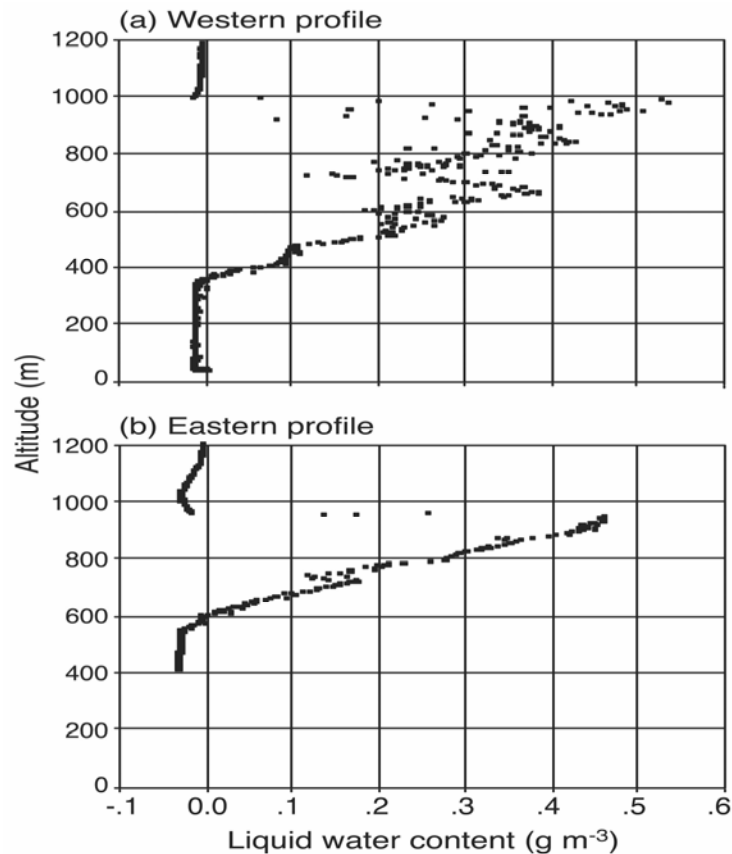
Several interesting findings came from this closure analysis. First, to get anything approaching good agreement between the model and observations, organics had to be included in the aerosol composition. Second, the distribution of organics both within particles and over the size distribution was important. The best agreement between model and observed values was obtained when solution activities were calculated using the Svenningsson model for such activities. This suggests the nonlinear impacts of organics on solution activity are the most important effects of organics on marine aerosol.

Some excellent data were also gathered during CARMA-III on the issue of mesoscale aerosol gradients and their impact on the mesoscale variability in cloud albedo as compared to the impact of other parameters. Examination of the horizontal gradients of cloud and subcloud properties on the mesoscale, together with simultaneously retrieved cloud albedo, has demonstrated that, while aerosol concentration certainly can modulate the cloud albedo on this scale, such properties as SST can have a similar impact and the convolution of the impacts of such factors can lead to either attenuation or enhancement of the aerosol effect alone (see Figure 4 for an example of re-enforcing effects). Additionally, analysis of vertical profiles of cloud properties suggests that aerosol impact on cloud albedo through modulation of cloud drop sedimentation can be either positive or negative as explained by the mechanism recently proposed by Ackerman et al (2004) and recent LES modeling. As a

consequence of this, the impact of aerosols via the second indirect effect (precipitation modulation) can be highly variable and, indeed, may be opposite to that previously proposed, i.e., higher aerosol concentrations can lead to thinner clouds with reduced albedo. An example of this sort of mechanism can be seen in Figure 5, where the liquid water path at the Eastern edge of a traverse has actually been substantially reduced by higher aerosol concentrations.



**Figure 4. Horizontal gradients observed on Flight 816 for (a) aerosol concentration, (b) sea surface temperature and (c) albedo**



**Figure 5. Liquid water vertical profiles for the (a) east and (b) west ends of the horizontal traverse from Flight 709**

## IMPACT/APPLICATIONS

These results demonstrate that the AHS provides a useful tool for exploring aerosol hygroscopicity. The hygroscopicity data as a function of aerosol size reveal that there are distinct patterns of DHGF for different aerosol type and illustrate the importance of chemical composition in the determination of aerosol hygroscopicity. The high but rather systematic variability in hygroscopicity over size suggests that mean aerosol hygroscopicity may be a poor predictor of the impact of this hygroscopicity on other aerosol integral properties or of the impact of aerosols on other processes, such as gas/aerosol interactions.

The results of the CCN closure analysis provide support for the substantial importance of organics in modulation of cloud drop number concentrations by aerosols. They furthermore suggest the main mechanism (chemical activity) by which this influence is exercised.

With respect to the relationship between aerosol and the albedo of overlying stratocumulus decks, our results support the importance of a number of different mechanisms, not merely aerosols, by which the CDNC can be modulated. Additionally, the analysis suggests that the impact of aerosols is not simple, and that even the sense (sign) of the aerosol forcing can change depending upon other variables. Hence, for example, purely meteorological factors cannot be neglected when assessing climate forcing by aerosols.

## RELATED PROJECTS

The size dependent hygroscopic growth of aerosols, including super-micron aerosols ( cf., Quinn et al, 1998), is a major factor in both the radiative energy balance of the lower marine atmosphere and the propagation of radiation through the MBL. Such radiation properties are necessary parameters for numerical modelers developing prognostic models. It is, furthermore, an aerosol characteristic closely related to CCN activity and, indeed, such activity can be predicted from it. Hence, these measurements are highly relevant to determination of CCN spectra and thus of the microphysics of MBL clouds.

The impact of both aerosol and SST gradients on cloud albedo is similarly important in attempting to predict MBL conditions since cloud cover will be strongly modulated by these factors.

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